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(73) Patenthaver: **Dräger Safety AG & Co. KGaA, Revalstrasse 1, 23560 Lübeck, Tyskland**

(72) Opfinder: **NAUBER, Andreas, Johannes Grevsen-Strasse 1, 23617 Stockelsdorf, Tyskland**
**SICK, Michael, Birkenallee 14, 23669 Timmendorfer Strand, Tyskland**
**MATTERN-FRÜHWALD, Marie-Isabell, Tremsbütteler Weg 88b, 22941 Bargteheide, Tyskland**
**CHRZAN, Rigobert, Ölmühle 10, 23843 Bad Oldesloe, Tyskland**
**SOMMER, Sabrina, Ginsterweg 12, 23558 Lübeck, Tyskland**
**METT, Frank, Schönböckener Strasse 3a, 23556 Lübeck, Tyskland**
**HENGSTENBERG, Andreas, Im Tannengrund 117, 23858 Reinfeld, Tyskland**
**STEINER, Gregor, Josef-Sorg-Straße 36, 79822 Titisee-Neustadt, Tyskland**

(74) Fuldmægtig i Danmark: **Chas. Hude A/S, H.C. Andersens Boulevard 33, 1780 København V, Danmark**

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# Liquid electrolyte for an electrochemical gas sensor

## Description

The invention pertains to a liquid electrolyte for an electrochemical gas sensor, especially for an electrochemical gas sensor for detecting $NH_3$ or gas mixtures containing $NH_3$.

Electrochemical gas sensors, with which the concentration of gaseous ammonia ($NH_3$) can be detected over a limited time period, are generally known. Such sensors are usually used in a great variety of industrial areas, ranging from the chemical industry to agricultural plants via the monitoring of refrigerating systems. They are used especially to recognize critical concentrations of the flammable ammonia gas, which is toxic and corrosive on inhalation, in good time and to warn against a corresponding risk.

The electrolyte used in the sensor is one of the essential components of such an electrochemical sensor. The electrolyte is in conductive contact with at least one anode and one cathode. If the gas to be detected enters the electrochemical sensor, a reaction, which leads to a measurable flow of current between the anode and the cathode of the sensor, will typically take place between the gas, the electrolyte and the material of the electrode.

Thus, EP 0 395 927 B1 describes an electrochemical measuring cell for determining ammonia or hydrazine in a gaseous or liquid test sample with at least one measuring electrode and one counterelectrode, which are accommodated in an electrolyte chamber filled with a soluble electrolyte, and which is closed by a permeable membrane towards the test sample.

EP 0 556 558 B1 also provides for an electrochemical measuring cell for determining ammonia, amines, hydrazine and hydrazine derivatives. It is proposed here that a hygroscopic alkali or alkaline earth salt be used as the conductive electrolyte in the electrolyte solution. This shall prevent the drying out of the

electrolyte and make possible in this way the most long-term usability possible of the sensor.

GB 2225859A discloses an electrolyte for determining ammonia. The electrolyte contains tris-(hydroxymethyl)-aminomethane hydrochloride and a hygroscopic additive selected from tetramethylammonium chloride or lithium chloride.

The detection of ammonia ($NH_3$) is carried out in electrochemical sensors of such a design by means of an electrochemical reaction between the ammonia gas flowing into the sensor, the electrodes and the electrolyte of the sensor. Entering ammonia gas is oxidized at the measuring electrode in the course of this reaction. The ammonium ions formed in the process are subsequently deprotonated again at the counterelectrode. However, it may prove to be problematic in this connection, for example, that additional nitrogen compounds may be formed as a byproduct of this reaction, which may lead to blocking (poisoning) of the electrode surfaces.

Based on this, an object of the present invention is to overcome these and other drawbacks of the state of the art.

To accomplish this object, the invention provides a liquid electrolyte corresponding to Claim 1. Embodiments are the subject-matter of the dependent claims.

In the case of a liquid electrolyte for an electrochemical gas sensor, especially for an electrochemical gas sensor that is suitable for the detection of $NH_3$ or $NH_3$-containing gas mixtures, the invention makes provisions for the electrolyte to contain at least one solvent, a conductive salt and/or an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof.

Especially for electrochemical gas sensors, in which electrodes consisting of noble metal or carbon nanotubes are used, such an electrolyte can be used with great advantage to improve the resistance of such a sensor to continuous gas admission. In particular, the risk of a poisoning, as was described above, can be markedly

minimized in this way.

According to the invention, the electrolyte contains a buffer corresponding to

Formula I $R^1$-($CR^2R^3$)$_n$-$SO_3H$

in which n = 1, 2, 3, 4 or 5, preferably n = 2 or n = 3, wherein all $R^2$ and $R^3$ are selected, independently from one another, from among H, NH and OH, and wherein $R^1$ is selected from the group containing piperazinyl, substituted piperazinyl, N-morpholino, cycloalkyl, and tris-(hydroxyalkyl)alkyl. For example, $R^2$ and $R^3$ may be selected, independently from one another, from among H, NH and OH, wherein n = 2 and $R^1$ is selected from the group containing piperazinyl, substituted piperazinyl, N-morpholino, cycloalkyl, and tris-(hydroxyalkyl)alkyl. It is also feasible, for example, that $R^2$ and $R^3$ are selected, independently from one another, from among H, NH and OH, wherein n = 2 and $R^1$ is selected from the group containing N-morpholino and tris-(hydroxyalkyl)alkyl. For example, it is especially advantageous here if n = 2 or n = 3, wherein all $R^2$ and $R^3$ are selected, independently from one another, from among H, NH and OH, and wherein $R^1$ is selected from among [4-(2-hydroxyethyl)-1]-piperazinyl, (N-morpholino), N-cyclohexyl, and tris-(hydroxymethyl)methyl. The buffer is especially preferably 3-(N-morpholino)-propanesulfonic acid or 3-(N-morpholino)-ethanesulfonic acid. It is thus conceivable, for example, that the electrolyte is a mixture of a solvent, a conductive salt and/or an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof, and wherein the electrolyte contains, in addition, a buffer, especially a buffer which is selected from among 3-(N-morpholino)-propanesulfonic acid or 3-(N-morpholino)-ethanesulfonic acid.

To prevent the electrolyte from drying out after a certain time, e.g. if the sensor shall be used in continuous operation, it is advantageous, moreover, if the electrolyte contains a component for lowering the vapor pressure as an additional component. The additional component may preferably be an alkylene glycol or polyalkylene glycol, and it is especially preferably propylene glycol, ethylene glycol or a mixture of propylene glycol and ethylene glycol. It is thus conceivable, for example, that the electrolyte is a mixture of a solvent, a conductive salt and/or

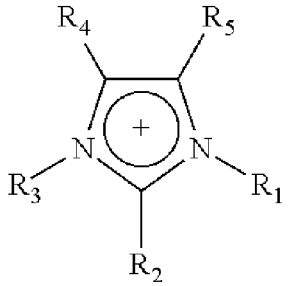
an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof, and wherein the electrolyte contains, moreover, at least one alkylene glycol, especially an alkylene glycol which is selected from among propylene glycol, ethylene glycol or a mixture of propylene glycol and ethylene glycol.

It is favorable, furthermore, if the solvent is selected from the group containing water and alkylene carbonate or mixtures thereof, preferably selected from the group containing water, propylene carbonate, ethylene carbonate or mixtures thereof. It is conceivable, for example, that the electrolyte is a mixture of a solvent, a conductive salt and/or an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof and wherein the solvent is water. As an alternative it is also feasible that the electrolyte is a mixture of a solvent, a conductive salt and/or an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof and wherein the solvent is alkylene carbonate, especially propylene carbonate, ethylene carbonate or a mixture of propylene carbonate and ethylene carbonate. In this connection, in particular, it is also feasible that the electrolyte is a mixture of a solvent, a conductive salt and/or an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof, wherein the electrolyte contains, moreover, a buffer, especially a buffer which is selected from among 3-(N-morpholino)-propanesulfonic acid or 3-(N-morpholino)-ethanesulfonic acid and wherein the solvent is alkylene carbonate, especially propylene carbonate, ethylene carbonate or a mixture of propylene carbonate and ethylene carbonate. It is moreover conceivable that the electrolyte is a mixture of a solvent, a conductive slat and/or an organic mediator, wherein the conductive salt is an ionic liquid, an inorganic salt, an organic salt or a mixture thereof, wherein the electrolyte contains, moreover, at least one alkylene glycol, especially an alkylene glycol which is selected from among propylene glycol, ethylene glycol or a mixture of propylene glycol and ethylene glycol, and wherein solvent is alkylene carbonate, especially propylene carbonate, ethylene carbonate or a mixture of propylene carbonate and ethylene carbonate.

The anion of the conductive salt is preferably selected from the group containing halides, carbonate, sulfonate, phosphate and/or phosphonate, preferably an anion selected from the group containing alkyl sulfonate, alkenyl sulfonate, aryl sulfonate, alkyl phosphate, alkenyl phosphate, aryl phosphate, substituted alkyl sulfonate, substituted alkenyl sulfonate, substituted aryl sulfonate, substituted alkyl phosphate, substituted alkenyl phosphate, substituted aryl phosphate, halogenated phosphate, halogenated sulfonate, halogenated alkyl sulfonate, halogenated alkenyl sulfonate, halogenated aryl sulfonate, halogenated alkyl phosphate, halogenated alkenyl phosphate, halogenated aryl phosphate, especially preferably an anion selected from the group containing fluorophosphate, alkyl fluorophosphate, aryl sulfonate, and especially preferably from the group containing perfluoroalkyl fluorophosphate and toluene sulfonate.

It is advantageous if the conductive salt contains metal ions, onium ions or a mixture of metal ions and onium ions as cations. For example, the metal ions may be selected from among alkali metal ions or alkaline earth metal ions, preferably from among Li, K and/or Na. It is favorable if the onium ions are selected from among ammonium, phosphonium, guanidinium cations and heterocyclic cations, preferably selected from among alkylammonium and heterocyclic cations, especially preferably selected from among alkylammonium, imidazolium and/or substituted imidazolium ions, wherein the substituted imidazolium ions preferably have a structure corresponding to:

Formula II




wherein R1, R2, R3, R4 and R5 may be selected, independently from one another, from among -H, straight-chain or branched alkyl containing 1 to 20 C atoms, straight-chain or branched alkenyl containing 2 to 20 C atoms and one or more double bonds, straight-chain or branched alkinyl containing 2 to 20 C atoms and one or more triple bonds, saturated, partially or fully unsaturated cycloalkyl

containing 3-7 C atoms, which may be substituted with alkyl groups containing 1 to 6 C atoms, saturated, partially or fully unsaturated heteroaryl, heteroaryl-C1-C6-alkyl or aryl-C1-C6-alkyl, wherein R2, R4 and R5 are especially preferably H, and R1 and R3 represent each, independently from one another, a straight-chain or branched alkyl containing 1 to 20 C atoms.

It is feasible in particular, for example, that tetrabutyl ammonium toluene sulfonate or 1-hexyl-3-methylimidazolium-tris(pentafluoroethyl)-trifluorophosphate is used as the conductive salt. As an alternative it is also feasible that the conductive salt is, for example, LiCl, KCl or a mixture of LiCl and KCl. It is thus especially advantageous if the electrolyte is a mixture of a solvent, a conductive salt and/or an organic mediator, wherein the conductive salt is selected from among LiCl, KCl, alkylammonium toluene sulfonate and ionic liquids, with a perfluoroalkyl fluorophosphate anion.

It is favorable, furthermore, if the organic mediator is a polyhydroxy compound, which forms a quinoid system or a naphthalene system during oxidation. For example, the organic mediator may be selected from the group containing ortho-dihydroxybenzene, para-dihydroxybenzene, substituted ortho-dihydroxybenzenes and substituted para-dihydroxybenzenes, dihydroxynaphthalene, substituted dihydroxynaphthalene, anthrahydroquinone, substituted anthrahydroquinone, preferably 1,2-dihydroxybenzene, 1,4-dihydroxybenzene, naphthohydroquinone, substituted 1,2- or 1,4-dihydroxybenzene, substituted hydroquinone, substituted naphthohydroquinone, especially preferably substituted anthrahydroquinone, substituted hydroquinone, and substituted 1,2-dihydroxybenzene. It is especially favorable in this connection if the substituents of the substituted anthraquinone, substituted 1,2-dihydroxybenzene and/or substituted 1,4-hydroquinone are selected from the group containing sulfonyl, tert-butyl, hydroxyl, alkyl, aryl, preferably sulfonic acid and/or tert-butyl.

It is especially favorable in any case if the electrolyte contains a mixture of propylene carbonate and/or ethylene carbonate as the solvent, contains LiCl, KCl, tetrabutylammonium toluene sulfonate and/or 1-hexyl-3-methyl-imidazolium

tris(pentafluoroethyl)-trifluorophosphate or a mixture of two or more of these components as the conductive salt and contains tert-butylhydroquinone and/or a substituted anthraquinone, preferably anthraquinone-2-sulfonate as organic mediator.

The concentration of the organic mediator may be between 10−6 mol/l and 10−2 mol/l. Thus, the organic mediator may be contained in the electrolyte at a concentration of $10^{-2}$ mol/l or less, preferably $10^{-3}$ mol/l or less, especially preferably $5 \cdot 10^{-4}$ mol/l or less, very especially preferably $2 \cdot 10^{-4}$ mol/l or less, and very especially preferably $10^{-4}$ mol/l or less. It is also feasible that the organic mediator is contained in the electrolyte at a concentration of $10^{-6}$ mol/l or more, preferably $10^{-5}$ mol/l or more, especially preferably $5 \cdot 10^{-5}$ mol/l or more, very especially preferably $8 \cdot 10^{-5}$ mol/l or more, and very especially preferably $10^{-4}$ mol/l or more. In particular, it is also feasible that the organic mediator is present at a concentration of $10^{-5}$ mol/l to $10^{-3}$ mol/l, preferably $5 \cdot 10^{-5}$ mol/l to $5 \cdot 10^{-4}$ mol/l, especially preferably $8 \cdot 10^{-5}$ mol/l to $2 \cdot 10^{-4}$ mol/l, and very especially preferably $10^{-4}$ mol/l.

An electrolyte according to the invention can be obtained especially preferably by means of a method that comprises the following steps:

a. Charging the solvent into a reaction vessel

b. Addition of the buffer

c. Addition of the organic mediator

d. Heating of the mixture while stirring for about 15 minutes at 150°C

e. Stirring for about one hour without further supply of heat until all solids are dissolved

f. Cooling to room temperature

g. Addition of the conductive salt

Further details and specifics appear from the figures described below and exemplary embodiments. In the drawings:

Fig. 1 is a schematic design of an electrochemical gas sensor, with which the electrolyte according to the invention for detecting ammonia can be used.

Fig. 2 is a schematic course of a detection reaction for $NH_3$ in an electrochemical gas sensor, which contains an electrolyte according to the invention.

Fig. 1 shows an electrochemical gas sensor 10, which has a housing 20 with an electrolyte reservoir 30. A gas inlet 21 and a gas outlet 22 are formed in the housing 20. A working electrode 51 is arranged within the housing 20 such that the working electrode 51 is in contact with gas that is flowing into the housing 20 through the gas inlet 21. The working electrode 51 is separated from a collecting electrode 52 by means of a glass fiber membrane 55. The collecting electrode 52 is in turn separated from the electrolyte reservoir 30 with a glass fiber membrane 55. Furthermore, a counterelectrode 53 and a reference electrode 54 are arranged within the electrolyte reservoir 30.

The electrolyte 40 according to the invention is present in the electrolyte reservoir 30. The glass fiber membranes 55 can be impregnated with the electrolyte. The electrolyte 40 can reach in this way both the working electrode 51 and the collecting electrode 52, so that a chemical reaction can take place there corresponding to the scheme shown in Fig. 2 between $NH_3$ flowing in, the material of the working and collecting electrodes 51, 52 and the electrolyte 40.

NH3 flowing into the gas sensor 10 reacts now on the surface of the working electrode 51 with the electrolyte. The working electrode 51 preferably consists, e.g., of a PTFE membrane with a carbon nanotubes coating. The counterelectrode 53 preferably consists of a noble metal. The electrolyte 40 is a composition of propylene carbonate and/or ethylene carbonate as the solvent, 1-hexyl-3-

methylimidazolium-tris(pentafluoroethyl)-trifluorophosphate as the conductive salt and tert.-butyl-1,2-dihydroxybenzene as the organic mediator in this example. The electrolyte preferably contains, furthermore, a buffer, namely 3-(N-morpholino)-propanesulfonic acid. As can be seen in Fig. 2, the tert.-butyl-1,2-dihydroxybenzene is oxidized into tert-butylquinone at the working electrode 51. The protons released in the process react with the NH3 flowing into the gas sensor 10 to form ammonium ions. The ammonium ions reach the counterelectrode 53, where the reverse reaction of the tert-butylquinone formed previously into 1,2-dihydroxybenzene takes place. $NH_3$, which can escape through the gas outlet 22, is released, in turn, from the ammonium ions. The buffer used stabilizes the pH value of the electrolyte, which is present between the working electrode and the counterelectrode 51, 53 in the electrolyte reservoir 30, in the course of this reaction process.

Exemplary embodiment for preparing an electrolyte according to the invention:

Polycarbonate is charged as a solvent into a reaction vessel. A 0.4-wt. % buffer, preferably 3-(N-morpholino)-propanesulfonic acid, is added to the polycarbonate. In the next step, 6.9 wt. % of the organic mediator, preferably tert-butyl-1,2-dihydroxybenzene, are added. The mixture is heated while stirring within 15 minutes, and a maximum temperature of 150°C is not exceeded. The mixture was subsequently stirred further for one hour without supplying more heat until all solids were dissolved. The solution obtained has a clear, slightly yellowish color.

The solution thus obtained is allowed to stand until it is cooled to room temperature. Then, 2.7 wt. % of the conductive salt, preferably Hmim-FAP (3-hexyl-3-methylimidazolium-tris(pentafluoroethyl)-trifluorophosphte), are added, and the mixture is stirred briefly, for about 1 minute.

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**List of references**

| | | | | | |
|---|---|---|---|---|---|
| | 10 | Gas sensor | | 40 | Electrolyte |
| | | | 10 | | |
| | 20 | Housing | | 51 | Working electrode |
| 5 | 21 | Gas inlet | | 52 | Collecting electrode |
| | 22 | Gas outlet | | 53 | Counterelectrode |
| | | | | 54 | Reference electrode |
| | 30 | Electrolyte reservoir | 15 | 55 | Glass fiber membrane |

## Patentkrav

**1.** Flydende elektrolyt til en elektrokemisk gassensor, især til en elektrokemisk gassensor der er egnet til påvisning af $NH_3$ eller $NH_3$-holdige gasblandinger, hvilken elektrolyt omfatter: mindst et opløsningsmiddel, et ledende salt og en organisk mediator, hvilket ledende salt er en ionisk væske, et uorganisk salt, et organisk salt eller en blanding deraf, **kendetegnet ved, at** elektrolytten omfatter en buffer, hvilken buffer omfatter en forbindelse

Formel I $R^1-(CR^2R^3)_n-SO_3H$

med n = 1, 2, 3, 4 eller 5, foretrukket n = 2 eller n = 3, hvor alle $R^2$ og $R.^3$ er uafhængigt valgt blandt H, NH og OH, og hvor $R^1$ er valgt fra gruppen, der omfatter: piperazinyl, substitueret piperazinyl, N-morpholino, cycloalkyl, tris (hydroxyalkyl) alkyl.

**2.** Elektrolytten ifølge krav 1, **kendetegnet ved, at** n = 2 eller n = 3, hvor alle $R^2$ og $R^3$ er uafhængigt valgt blandt H, NH og OH, og hvor $R^1$ er valgt blandt [4-(2-hydroxyethyl)-1]-piperazinyl, (N-morpholino), N-cyclohexyl, tris-(hydroxymethyl) methyl, hvor bufferen fortrinsvis er 3-(N-morpholino)propansulfonsyre eller 3 -(N-morpholino)ethansulfonsyre.

**3.** Elektrolyt ifølge et af kravene 1 og 2, **kendetegnet ved, at** elektrolytten som en yderligere komponent omfatter en komponent til at sænke damptrykket, hvilken yderligere komponent fortrinsvis er en alkylenglycol eller polyalkylenglycol, mere foretrukket propylenglycol, ethylenglycol eller en blanding af propylenglycol og ethylenglycol.

**4.** Elektrolyt ifølge mindst et af kravene 1 til 3, **kendetegnet ved, at** opløsningsmidlet er valgt fra gruppen, der omfatter: vand og alkylencarbonat eller blandinger deraf, fortrinsvis er valgt fra gruppen, der omfatter: vand, propylencarbonat, ethylencarbonat eller blandinger deraf.

**5.** Elektrolyt ifølge mindst et af kravene 1 og 4, **kendetegnet ved, at** anionen af det ledende salt er valgt fra gruppen, der omfatter: halogenider, carbonat, sulfat, phosphat og/eller phosphonat.

**6.** Elektrolyt ifølge mindst et af kravene 1 til 5, **kendetegnet ved, at** det ledende salt som kationer indeholder metalioner, oniumioner eller en blanding af metalioner og oniumioner.

**7.** Elektrolytten ifølge krav 6, **kendetegnet ved, at** metalionerne er valgt blandt alkalimetalioner og jordalkalimetalioner, fortrinsvis fra Li, K og/eller Na.

**8.** Elektrolyt ifølge et af kravene 6 og 7, **kendetegnet ved, at** oniumionerne er valgt blandt ammoniumkationer, phosphoniumkationer, guanidiniumkationer og heterocykliske kationer.

**9.** Elektrolyt ifølge et hvilket som helst af kravene 1 til 8, **kendetegnet ved, at** den organiske mediator er en polyhydroxylforbindelse, der danner et quinoidsystem eller et naphthalensystem ved oxidation.

**10.** Elektrolytten ifølge krav 9, **kendetegnet ved, at** den organiske mediator er valgt fra gruppen, der omfatter: ortho-dihydroxybenzen, para-dihydroxybenzen, substitueret ortho-dihydroxybenzen og substitueret para-dihydroxybenzen, dihydroxynaphthalen, substitueret dihydroxynaphthalen, anthrahydroquinon, substitueret anthrahydroquinon.

**11.** Elektrolytten ifølge krav 10, **kendetegnet ved, at** substituenterne i den substituerede anthraquinon, substituerede 1,2-dihydroxybenzen og/eller substituerede 1,4-hydroquinon er valgt fra gruppen, der omfatter: sulfonyl, tert-butyl, hydroxyl, alkyl, aryl, fortrinsvis sulfonsyre og/eller tert-butyl.

**12.** Elektrolyt ifølge et hvilket som helst af kravene 1 til 11, **kendetegnet ved, at** elektrolytten omfatter en blanding af propylencarbonat og/eller ethylencarbonat som opløsningsmiddel, LiCl, KCl, tetrabutylammoniumtoluensulfonat og/eller 1-

hexyl-3-methylimidazolium tris(pentafluorethyl)trifluorphosphat eller en blanding af to eller flere af disse komponenter som ledende salt, og tert-butylhydroquinon og/eller en substitueret anthraquinon, fortrinsvis anthraquinon-2-sulfonat, som organisk mediator.

**13.** Elektrolyt ifølge et hvilket som helst af kravene 1 til 12, **kendetegnet ved, at** den organiske mediator er til stede i elektrolytten i en koncentration på $10^{-2}$ mol/l eller mindre, fortrinsvis på $10^{-3}$ mol/l eller mindre, mere foretrukket på $5*10^{-4}$ mol/l eller mindre, endnu mere foretrukket på $2*10^{-4}$ mol/l eller mindre, endnu mere foretrukket på $10^{-4}$ mol/l eller mindre.

**14.** Elektrolyt ifølge et hvilket som helst af kravene 1 til 13, **kendetegnet ved, at** den organiske mediator er til stede i elektrolytten i en koncentration på $10^{-6}$ mol/l eller mere, fortrinsvis på $10^{-5}$ mol/l eller mere, mere foretrukket på $5*10^{-5}$ mol/l eller mere, endnu mere foretrukket på $8*10^{-5}$ mol/l eller mere, endnu mere foretrukket på $10^{-4}$ mol/l eller mere.

**15.** Fremgangsmåde til fremstilling af en elektrolyt ifølge et hvilket som helst af de foregående krav der omfatter trinnene:

a. initialt at indføre opløsningsmidlet i et reaktionskar
b. at tilføje bufferen
c. at tilføje den organiske mediator
d. at opvarme blandingen til 150 °C under omrøring i ca. 15 minutter
e. at omrøre i cirka en time uden yderligere varmeforsyning, indtil alle faste stoffer er opløst
f. at afkøle til stuetemperatur, og
g. at tilsætte det ledende salt.

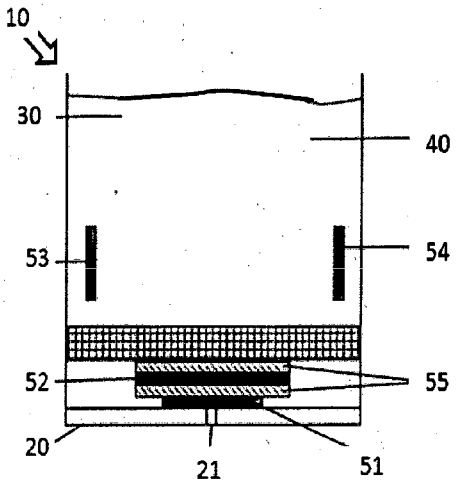
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Fig. 1

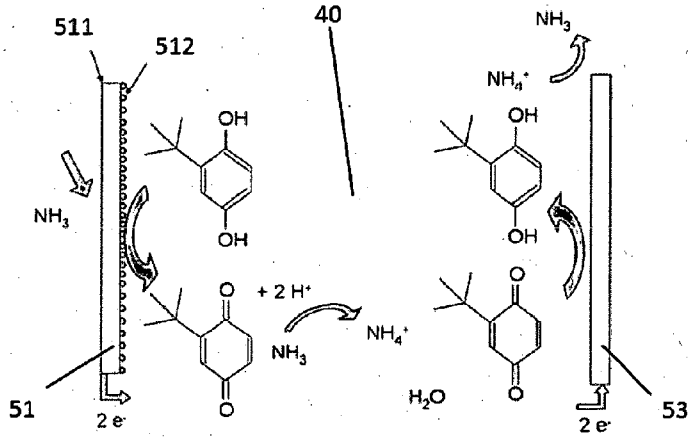
511
512
40
NH3
NH4+
OH
OH
NH3
+ 2 H+
NH4+
NH3
O
O
H2O
O
51
2 e-
2 e-
53

Fig. 2